

Short-time β -relaxation in glass-forming liquids is cooperative in nature

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Temporal relaxation of density fluctuations in supercooled liquids near the glass transition occurs in multiple steps. The short-time β -relaxation is generally attributed to spatially local processes involving the rattling motion of a particle in the transient cage formed by its neighbors. Using molecular dynamics simulations for three model glass-forming liquids, we show that the β -relaxation is actually cooperative in nature. Using finite-size scaling analysis, we extract a growing length-scale associated with β -relaxation from the observed dependence of the β -relaxation time on the system size. Remarkably, the temperature dependence of this length scale is found to be the same as that of the length scale that describes the spatial heterogeneity of local dynamics in the long-time α -relaxation regime. These results show that the conventional interpretation of β -relaxation as a local process is too simplified and provide a clear connection between short-time dynamics and long-time structural relaxation in glass-forming liquids.

I. INTRODUCTION

Temporal relaxation of density fluctuations in supercooled liquids near the glass transition occurs in multiple steps. At short times, the temporal auto-correlation of density fluctuations and related correlation functions approach a plateau after a fast initial decay. This part of the relaxation is known as the β relaxation. The subsequent long-time decay of the correlation functions from the plateau to zero is known as the α relaxation. The origin of this non-exponential, multi-step relaxation and possible connections between the short-time β relaxation and the long-time α relaxation are among the fundamental issues in the study of glassy dynamics.

Many approaches to understanding slow relaxation in glass forming liquids invoke the notion of a growing length scale that governs the increase of relaxation time scales [1–9]. Experimental and theoretical studies of dynamic heterogeneity in glass-forming liquids [10–13] have lead to a detailed analysis of length scales that are associated with spatial correlations of the mobility of particles [14–18]. These correlations have been studied through a four-point density correlation function $g_4(r, t)$ [19–21], its Fourier transform - the four-point structure factor $S_4(q, t)$ - and the associated dynamic susceptibility $\chi_4(t) \equiv \lim_{q \rightarrow 0} S_4(q, t)$ [8, 9, 14, 22, 23]. Analytic predictions for the behaviour of dynamic length scales and susceptibilities in both short-time (β) and long-time (α) relaxation regimes have been obtained from inhomogeneous mode coupling theory (IMCT) [3, 14, 15]. These predictions include an initial power-law growth of the cooperativity length scale in time in the β relaxation regime, followed by a saturation at time scales comparable to the α relaxation time. However, details of how the dynamics crosses over from its short-time behaviour to that at long times and the correspond-

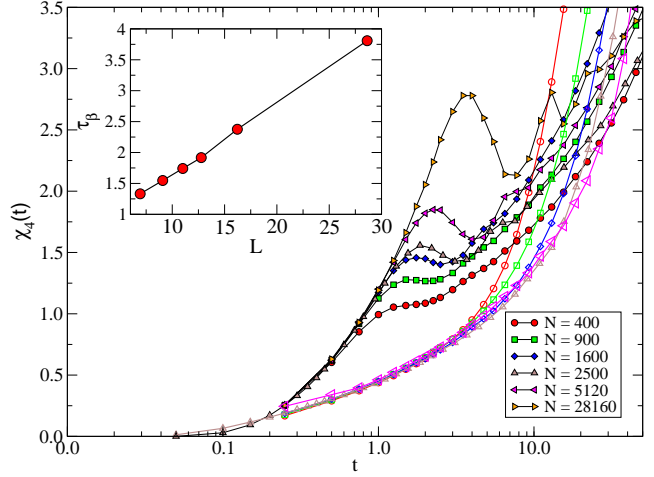


FIG. 1. Four-point susceptibility $\chi_4(t)$ for different system sizes, plotted versus time t . Note the appearance of a peak at short times. The height of the peak increases with increasing system size. The filled symbols represent data from molecular dynamics simulations and open symbols show data from Monte Carlo Simulations. The peak of χ_4 at short time scales disappears in Monte Carlo simulations. Inset: The height of the short-time peak of χ_4 is plotted as a function of the length L of the sample. The height is found to increase linearly with L .

ing crossover in the dynamic length scale(s) are still not well-understood. There are many ideas and observations that attempt to relate dynamical features observed at short time scales to long-time structural relaxation [24–31]. Thus, a proper understanding of relaxation processes in both β and α regimes and their underlying relation (if any) is extremely important in the overall understanding of glassy dynamics and its rich phenomenology.

In an earlier study [34], the method of finite-size

scaling (FSS) [32], used extensively for obtaining accurate numerical results for critical properties near conventional phase transitions, was used to obtain the length scale $\xi(T)$ associated with dynamical heterogeneity, by analyzing the size dependence of the dynamic susceptibility $\chi_4(t)$ and the associated Binder cumulant. It was subsequently shown that the heterogeneity length obtained in this way is in good agreement with that obtained by analyzing the four-point structure factor $S_4(q, t)$ [35, 36]. However, very large system sizes needed to be employed in the analysis of $S_4(q, t)$, and further difficulties arise from the ensemble dependence of the associated susceptibility $\chi_4(t)$ [37, 38]. Indeed, results [39] claiming to confirm IMCT predictions for $\chi_4(t)$ and the associated length scale $\xi(T)$ are affected by finite-size effects. Thus, notwithstanding caveats about its use [37, 40], FSS offers an attractive approach to study length scales relevant to glassy dynamics. We employ this method in the present work.

It was found, however, in [34] that the α relaxation time τ_α that describes the long-time decay of a two-point density correlation function does not exhibit the expected dynamical scaling, in that (a) the time scale decreases instead of increasing with system size, and (b) a plot of τ_α scaled by its asymptotic value for large system sizes *vs.* similarly scaled values of χ_4 does not display a scaling collapse. This was interpreted as indicating a mixing of activated and non-activated mechanisms of structural relaxation (see also [41, 42]). It has been suggested [42] that behaviour described by IMCT, without the influence of activated dynamics, should be expected instead in the short-time dynamics, in a temperature window above the mode coupling transition temperature.

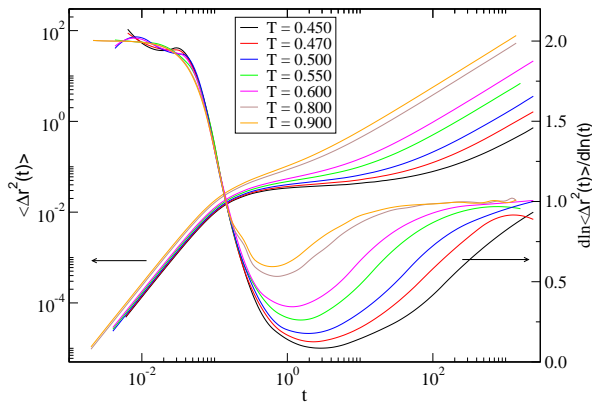


FIG. 2. Mean square displacement (MSD) and the derivative of the logarithm of MSD with respect to the logarithm of time t , shown as a function of t . The minimum of the derivative defines τ_β . (see text for details).

In this article, we describe the results of FSS stud-

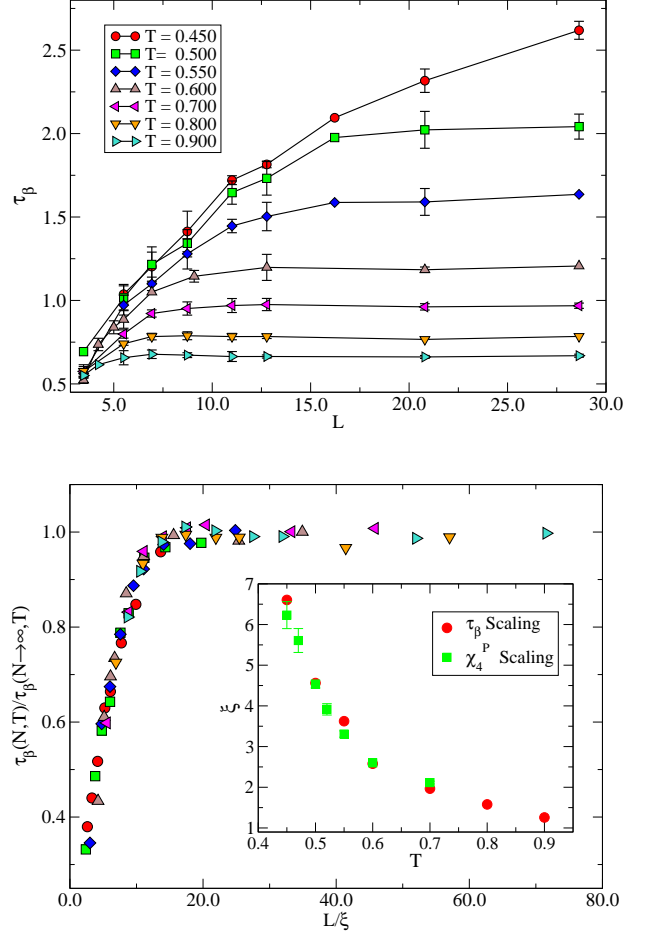


FIG. 3. Top panel: τ_β for the 3dKALJ model, shown as a function of system size for different temperatures in the interval $T \in [0.45, 0.90]$. τ_β increases with increasing system size, saturating at a value that increases with decreasing temperature. Bottom panel: Scaling collapse of τ_β . For each temperature, the linear size L is scaled by a length $\xi(T)$ such that data for all temperatures collapse to a master curve. Inset: Comparison of the correlation length extracted from the system-size dependence of τ_β with that obtained from the α regime [34] as a function of temperature. The two length scales agree with each other to a good accuracy.

ies of a short time scale (the β relaxation time scale τ_β) for three generic model glass-formers. The simulation details are provided in Sec. II. We compare the estimated length scales with those obtained from FSS analysis of χ_4^P , the peak value of $\chi_4(t)$, which quantifies dynamic heterogeneity at the α relaxation time scale, and find that the two length scales agree well for all the systems we study. We also find a power-law dependence of the β time scale on the length scale, in qualitative agreement with the prediction of IMCT. These results are described in Sec. III. We also perform an analysis of the dependence of the behaviour

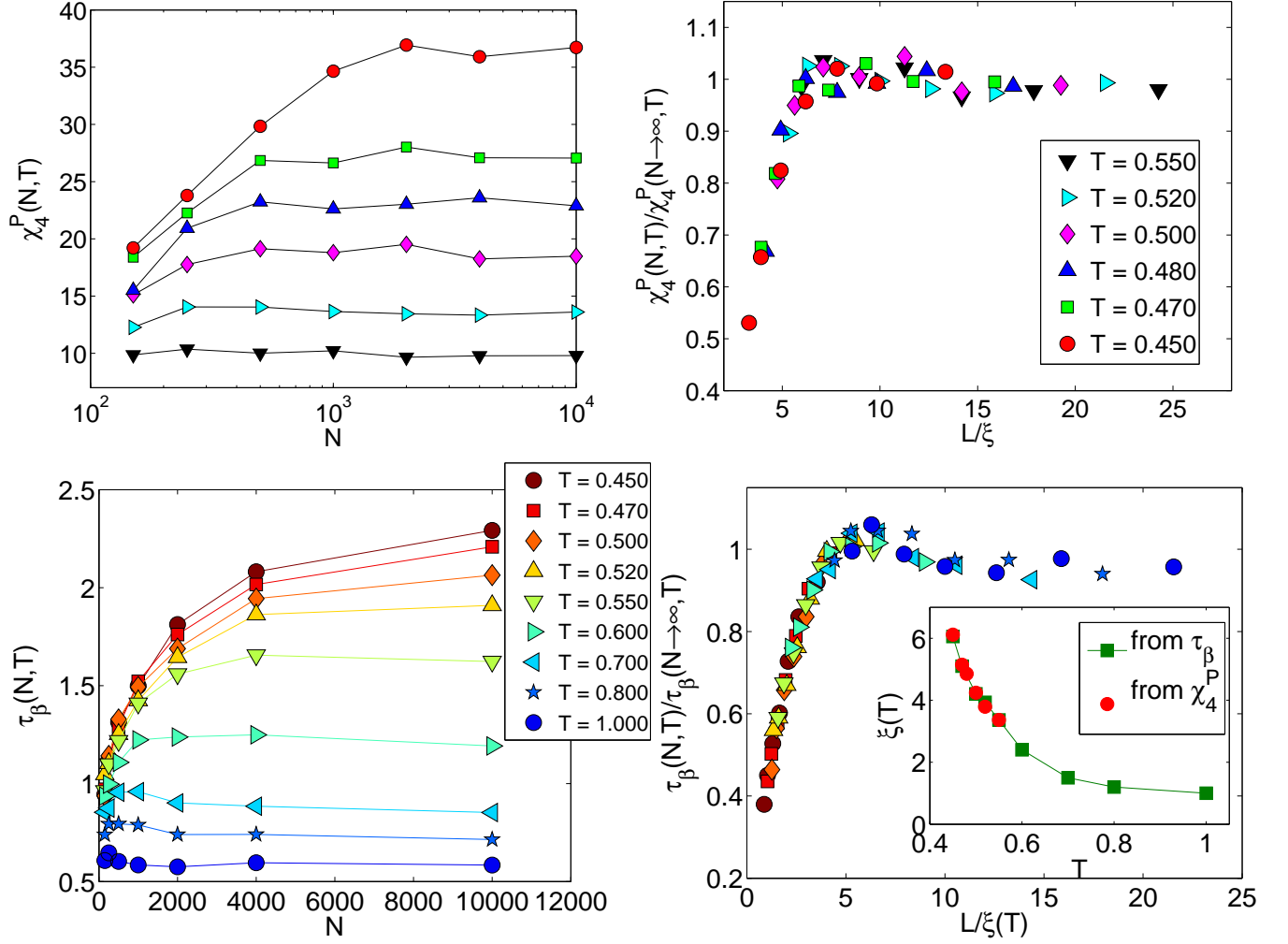


FIG. 4. Top left panel: System-size dependence of χ_4^P , the peak value of $\chi_4(t)$, for different temperature for the 3dIPL model system. Top right panel: The data collapse of $\chi_4^P(N, T)$ to extract the dynamic heterogeneity length scale. $\chi_4^P(N \rightarrow \infty, T)$ is the asymptotic value of χ_4^P in the limit of infinite system size. The corresponding length scales are shown in the inset of the bottom right panel of this figure (red circles). Bottom left panel: System-size dependence of τ_β for all the temperature studied, including high temperatures. Bottom right panel: Data collapse of $\tau_\beta(N, T)$ to obtain the length scale associated with the cooperativity in β -relaxation. Inset: Comparison of the correlation length extracted from the system-size dependence of τ_β with that obtained from the system-size dependence of χ_4^P , as a function of temperature. The two length scales agree with each other to a good accuracy.

in the β regime on the microscopic dynamics, as described in Sec. IV. Sec. V contains a discussion of our results and a summary.

II. METHOD AND SIMULATION DETAILS

We study three model liquids in this work. They are: (i) the three-dimensional Kob-Andersen binary Lennard-Jones mixture (referred here as 3dKALJ) [44], (ii) a three-dimensional system characterized by a repulsive inverse power-law potential (3dIPL) considered in Ref. [45] and (iii) a three-dimensional sys-

tem characterized by a repulsive inverse power-law potential (3dR10) [46] whose range of interaction is smaller compared to that in the first two models. For the first two model systems the interaction range covers the second neighbouring shell whereas for the third case it is truncated within the first shell.

We have performed a series of simulations, for systems of sizes ranging from $N = 150$ to $N = 28160$ particles at number density $\rho = 1.20$ of the glass-forming Kob-Andersen binary liquid mixture [44]. Details of the model parameters and the reduced units used for length, time and temperature are the same as in [34]. We have done simulations for 7 different tem-

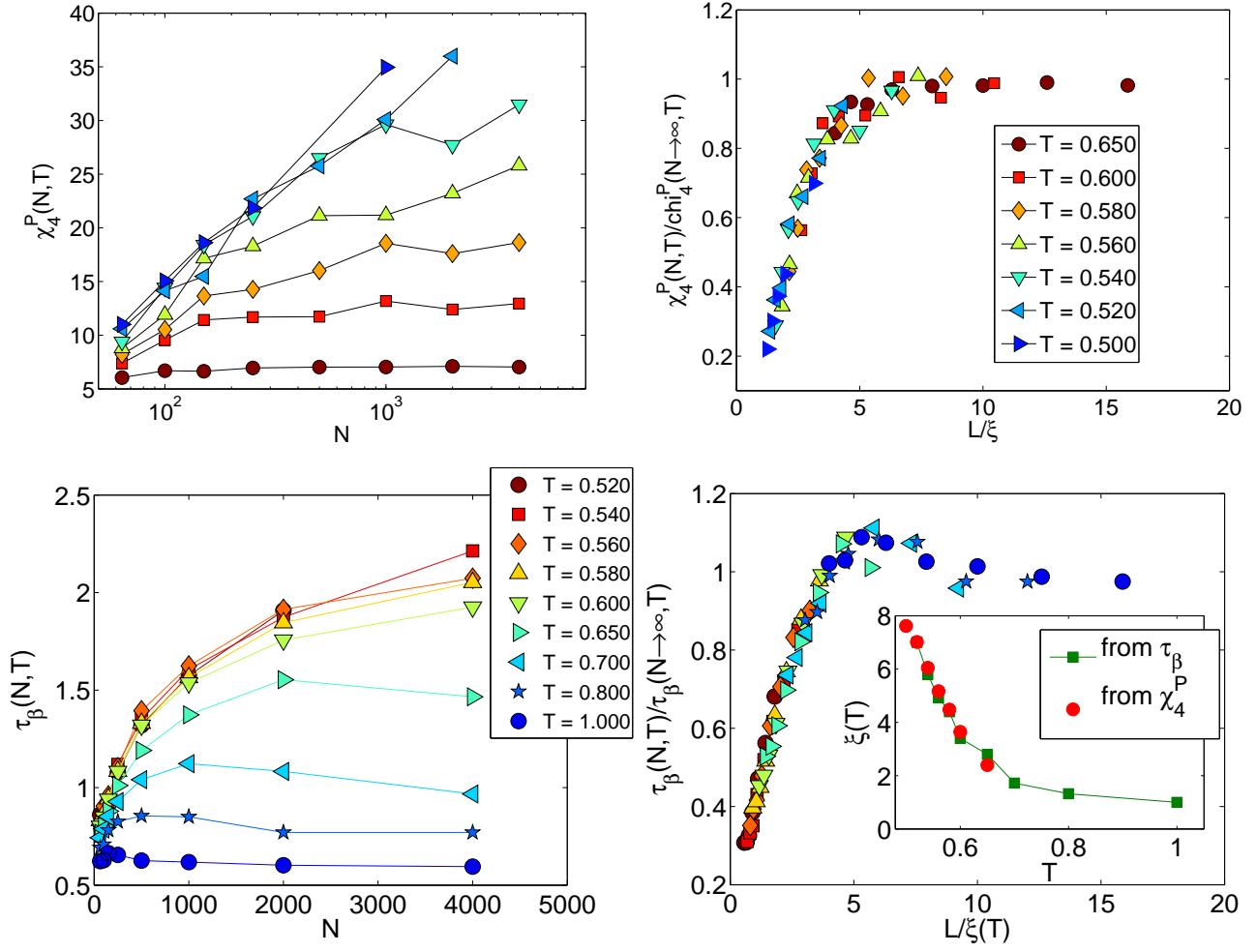


FIG. 5. Top left panel: System-size dependence of χ_4^P , the peak value of $\chi_4(t)$, for different temperature for the 3dR10 model system. Top right panel: The data collapse of $\chi_4^P(N, T)$ to extract the dynamic heterogeneity length scale. $\chi_4^P(N \rightarrow \infty, T)$ is the asymptotic value of χ_4^P in the limit of infinite system size. The corresponding length scales are shown in the inset of the bottom right panel of this figure (red circles). Bottom left panel: System-size dependence of τ_β for all the temperature studied, including high temperatures. Bottom right panel: Data collapse of $\tau_\beta(N, T)$ to obtain the length scale associated with the cooperativity in β -relaxation. Inset: Comparison of the correlation length extracted from the system-size dependence of τ_β with that obtained from the system-size dependence of χ_4^P , as a function of temperature. The two length scales agree with each other to a good accuracy.

peratures in the range $T \in [0.900, 0.450]$. For the 3dIPL model the system sizes studies are in the range $N \in [150, 10000]$ at number density $\rho = 1.20$ for 9 different temperatures in the range $T \in [0.450, 1.000]$. For the 3dR10 model system sizes studied are in range $N \in [64, 40000]$ at number density $\rho = 0.81$ for 9 temperatures in the range $T \in [0.50, 1.000]$. All the simulations are done in the canonical ensemble using a modified leap-frog integration scheme with the Berendsen thermostat. We have also performed simulation with another constant temperature simulation algorithm due to Brown and Clark [47]. The results do not depend on the exact algorithm used for integrating the equations of motion. The equilibration runs for all these temperatures are close to 100τ

(where τ is the α -relaxation time estimated from the decay of the two-point density correlation function) and we have averaged the data over 32 independent runs of length 100τ . For the results discussed in Sec. IV, we performed Brownian Dynamics simulations using the predictor-corrector algorithm given in [48].

In [34], the α relaxation time was identified by the decay of a two-point density correlation function, as well as by considering the location of the peak of $\chi_4(t)$. In order to define a β time scale, we first consider an analogous procedure of locating a short-time feature in $\chi_4(t)$. Indeed, as observed in [43], there exists a maximum in $\chi_4(t)$ at short times, whose location and height depend on the system size, as shown

in Fig. 1. However, we find that the identified time scale increases linearly with the length L of the system. This behaviour is shown in the inset of Fig. 1. This plot also shows that this time scale continues to increase with increasing L without showing any sign of saturation. These observations suggest a phononic origin of this time scale. Such a possibility is further supported by the fact that this feature disappears when we use Monte Carlo simulations to calculate $\chi_4(t)$. We opt not to use this procedure for identifying τ_β .

In [39], a short time scale τ_β was identified as the time at which an inflection occurs in a log-log plot of the mean squared displacement *vs.* time. We use the same definition to evaluate τ_β as a function of system size and temperature. In Fig. 2, we show the mean squared displacement (MSD) as a function of time in a log-log plot and its derivative. The clear dip in the derivative and its variation with temperature indicate that this estimation of τ_β can be done unambiguously and without much uncertainty. We use this definition of τ_β in the present work.

III. RESULTS

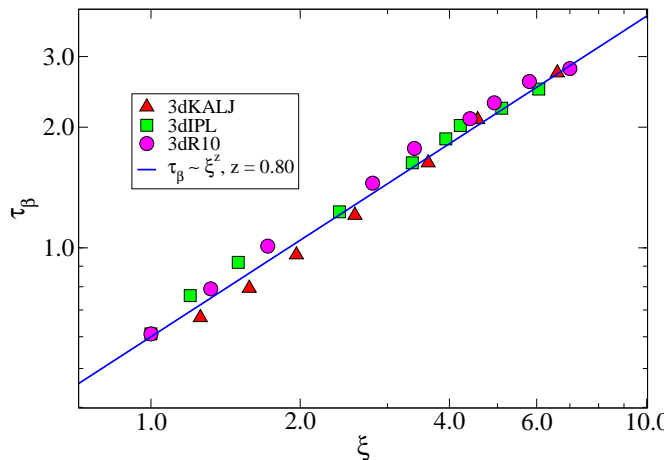


FIG. 6. The relaxation time τ_β , plotted as a function of the correlation length ξ , exhibits a power-law dependence, albeit over a small range of values. The line drawn through the data points represents a power-law with exponent $z = 0.80$.

In Fig. 3, we show the system-size dependence of τ_β for different temperatures for the 3dKALJ model. For high temperatures the dependence is weak and the asymptotic value is reached for small system sizes, but this characteristic size becomes increasingly large with decreasing temperature. Thus, τ_β shows a size dependence that is opposite to that displayed by τ_α [34], exhibiting a system-size dependence that may

normally be expected for a time scale determined by an underlying length scale in the system. We note that the range over which τ_β varies with system size and temperature is modest, unlike the behaviour observed for τ_α . Another important observation about the system-size dependence of τ_β is that the dependence of this time scale on the system size becomes negligibly small if the system size is sufficiently large. This is very different from the behaviour of the time scale associated with the short-time peak of $\chi_4(t)$ shown in Fig. 1. As shown there, this time scale, which we attribute to phonon-like excitations, continues to increase linearly with the length L of the system without showing any sign of saturation at large values of L .

We next scale the linear system size L at each temperature by an empirically determined length $\xi(T)$ such that the values of $\tau_\beta(N, T)/\tau_\beta(N \rightarrow \infty, T)$ for all N and T ($\tau_\beta(N \rightarrow \infty, T)$ is the large- N asymptotic value of $\tau_\beta(N, T)$) collapse onto a master curve when plotted versus $L/\xi(T)$. The data collapse obtained this way, shown in Fig. 3, is very good. This allows for the determination of a β -regime length scale $\xi(T)$.

We then compare the length scale estimated in this way with the heterogeneity length estimated in [34] from quantities obtained at τ_α . This comparison is shown in the inset of the bottom panel of Fig. 3. In order to make the comparison, we scale the present estimate so that the two length scales match at $T = 0.6$. Length scales obtained by FSS are known only up to a multiplicative factor, and hence this procedure does not introduce any additional arbitrariness. We find that the temperature dependence of the two length scales agrees very well. IMCT predicts that ξ grows as a power law in time up to τ_β and stays at the value τ_β up to τ_α . Our results therefore confirm this expectation. Independently of IMCT predictions, the agreement is remarkable in pointing to an intimate connection between dynamics at short times (β regime) and long times (α regime). This result suggests that the heterogeneity present in the dynamics in the α regime has been built up already in the β regime, and therefore in principle, essential information about the α relaxation can be obtained by studying short-time dynamics. Our result therefore lends support to many investigations and ideas that aim to relate dynamical behaviour at short times to long-time structural relaxation [24–31].

To see whether the results obtained for the 3dKALJ model are generic, we have done similar analysis for the 3dIPL and 3dR10 models. In Fig. 4, we have shown, in the top left panel, the system-size dependence of the peak value of the four-point susceptibility for different temperatures for the 3dIPL model. The top right panel shows the results of FSS performed for the same data to obtain the dynamic heterogeneity length scale. In the bottom left panel we show the system-size dependence of the short time scale τ_β for different temperatures and in the bottom right panel

the corresponding FSS of τ_β . The scaling collapse observed in this case is also very good. We find that the temperature dependence of the length scale obtained from the FSS of τ_β matches quite well with the dynamic heterogeneity length scale, as shown in the inset of the bottom right panel of Fig.4. Similar analysis done for the 3dR10 model confirms that the same observations hold for this model too, as shown in Fig.5.

Finally we consider the dependence of τ_β on the extracted length scale ξ . In Fig. 6, we show the relaxation time as a function of the extracted correlation length in a log-log plot. In contrast to τ_α which exhibits deviations from a power-law dependence on the corresponding length scale [34], we find that a power law relation $\tau \sim \xi^z$, holds for τ_β . Although such a dependence is in qualitative agreement with IMCT, we find $z \simeq 0.80$ which is at variance with the IMCT prediction[3].

IV. DEPENDENCE ON MICROSCOPIC DYNAMICS

In this section we present results concerning the dependence of the time scale τ_β on the details of the microscopic dynamics. To address this issue systematically, we have performed Brownian dynamics (BD) simulations using a predictor-corrector scheme [48]. This simulation scheme allows one to change systematically the friction coefficient to go from the very low friction limit (close to a molecular dynamics simulation) to overdamped dynamics with large friction (close to a Monte Carlo simulation). We have changed the friction in our simulations over one order of magnitude and studied its effect on the β -relaxation time scale. We find that although a β time scale can be identified unambiguously for all the values of the friction coefficients studied, the system-size dependence becomes weaker with increasing friction and almost completely goes away for the largest value of the friction coefficient considered here. The system-size dependence of τ_β is shown in Fig.7 for the 3dKALJ model at temperature $T = 0.470$ for system sizes in the range $N \in [150, 10000]$. However, it is important to note that the time scale itself remains well-defined with increasing friction. This is different from the behaviour of the time scale obtained from the short-time peak in $\chi_4(t)$ [43]: as discussed above, this time scale can not be defined for Monte Carlo dynamics because the short-time peak in $\chi_4(t)$ is not present for this dynamics. Our results are consistent with previous work comparing the results for different microscopic dynamics [49], which found that the behaviour in the early β regime is affected by the microscopic dynamics.

We do not have a full understanding of the observed influence of the microscopic dynamics on the system-size dependence of τ_β . There are reasons to

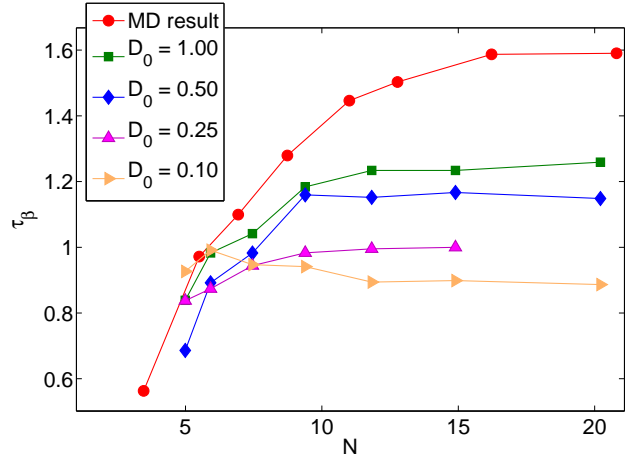


FIG. 7. System-size dependence of the short-time β -relaxation time scale τ_β at $T = 0.470$ for the 3dKALJ model glass former from Brownian dynamics simulations with different values of the friction coefficient. D_0 indicated in the legend is the microscopic diffusivity of the particles and is inversely proportional to the damping coefficient. Notice that for small damping (large D_0) the system-size dependence is very similar to that obtained in molecular dynamics simulations. The system-size dependence becomes weaker with increasing damping. The time scale for Brownian dynamics is scaled to match the long-time mean squared displacement for both molecular dynamics and Brownian dynamics.

believe that the observed behaviour is connected to the effects of the properties of the inherent structures (local minima of the potential energy), whose basins of attraction are visited by the system during its time evolution, on its dynamics. The dynamics in the short-time β -relaxation regime at low temperatures is expected to be strongly influenced by the properties of the inherent structures because the system should remain confined in the basin of a single inherent structure (or in a single metabasin [50], depending on the temperature) during its evolution over relatively short times. A recent study [51] has shown that the dynamics of the 3dKALJ model in the β relaxation regime (up to time scales that are relatively short, but longer than the τ_β considered in our work), observed in molecular dynamics simulations at temperatures near and below the glass transition temperature T_c of mode coupling theory ($T_c \simeq 0.435$ for the 3dKALJ model), can be understood from the low-energy properties of the relevant inherent structures. These properties include the eigenvalues and eigenvectors of the Hessian matrix evaluated at the potential energy minimum (these define the “normal modes” of small-amplitude oscillations near the bottom of the basin of an inherent structure) and the third and fourth derivatives of the potential energy at the minimum (these coefficients determine the effects

of anharmonicity on the normal modes). Thus, the system-size dependence of τ_β obtained in our Newtonian molecular dynamics simulations and the length scale we have extracted from this dependence should be closely related to these properties of the inherent structures. Our observation that the length scale extracted from the system-size dependence of τ_β obtained from molecular dynamics simulations is essentially the same as the length scale of dynamic heterogeneity at the α -relaxation time scale then suggests that the spatial structure of dynamic heterogeneity at time scales of the order of the α -relaxation time is closely related to the aforementioned properties of the inherent structures. This suggestion receives strong support from several experimental [52] and numerical [27, 53, 54] studies that have shown that the spatial structure of dynamic heterogeneity at the α -relaxation time scale is closely related to the structure of the eigenvectors associated with some of the low-lying eigenvalues of the Hessian matrix evaluated at the appropriate inherent structures. All these observations suggest that the short-time (β -relaxation) and long-time (α -relaxation) dynamics observed in molecular dynamics simulations are closely related to each other through the low-energy properties of the relevant inherent structures. In Brownian dynamics simulations with a large friction coefficient, the influence of the properties of the inherent structures on the short-time dynamics is masked by the strong damping of the normal modes. This may reduce or completely eliminate some of the effects of the structural properties of the inherent structures on the dynamics. The system-size dependence of τ_β which, as argued above, is closely related to the low-energy properties of the inherent structures, may be one of the effects that disappear in the presence of strong friction. These arguments provide a rationalization of the observed effects of strong friction on the system-size dependence of τ_β obtained from Brownian dynamics simulations and suggest that the low-energy properties of the inherent structures visited by the system during its time evolution play an important role in both β and α relaxation processes.

V. DISCUSSION

In this work, we have studied the system-size dependence of the β relaxation time for three model

liquids in three dimensions. In each case, we find that the time scale initially increases with increasing system size and saturates for large values of the system size, exhibiting behaviour conforming to usual expectations for the size dependence of a quantity that depends on a length scale. This is unlike the system-size dependence of the α relaxation time that is found to decrease with increasing system size [34]. On the other hand, the length scale extracted from FSS of this β relaxation time scale matches very well with the length scale extracted from the FSS of the four-point susceptibility $\chi_4^P(T)$ measured at the α -relaxation time scale. This is in agreement with expectations based on inhomogeneous mode coupling theory, as also our observation that the asymptotic value of the β time scale, obtained for large systems, exhibits a power-law dependence on the length scale. The value of exponent of this dependence, however, does not match IMCT predictions ($\frac{4}{2a} \simeq 5.4$) [3]. Our results are also consistent with ideas that relate dynamics at short times to long-time structural relaxation [24–31]. We have also presented intriguing results for the dependence of the short-time dynamics on the microscopic dynamics used in the simulation and suggested a possible explanation of the observed behaviour in terms of the role played by certain low-energy properties of the relevant inherent structures (i.e. inherent structures whose basins are visited by the system during its time evolution) in the dynamics of the system.

The results reported here have a strong connection with those in [55], in which oscillatory shear simulations were performed to study the effects of short-time β -relaxation in a supercooled two-dimensional glass-forming liquid at both high and low temperatures. The loss modulus measured in this study was shown to be related to β -relaxation and the effect of this relaxation was found to decrease sharply by the introduction of a small amount of pinning disorder in the system. The cooperative displacements of individual particles over the β -relaxation time scale were found to diminish very rapidly with the introduction of pinning disorder. Based on these observations, it was concluded that β -relaxation is also cooperative in nature, similar to α -relaxation. Our study clearly shows that β -relaxation is indeed cooperative and the cooperativity is likely to have the same origin as that in α -relaxation.

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